

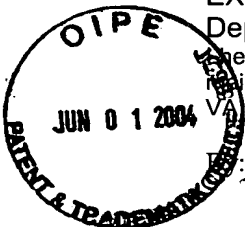
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To: Judith Muzyk Date: June 1, 2004  
Judith Muzyk

Docket No.: 9913/9-1556

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Johan C. FITTTER Conf. 6321  
Serial No.: 09/927,805 Group Art Unit: 1746  
Filing Date: August 10, 2001 Examiner: Bruce F. Bell  
For: ELECTROCHEMICAL CELLS AND AN INTERCHANGEABLE  
ELECTROLYTE THEREFORE

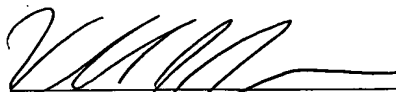
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

**SUBMISSION OF PRIORITY DOCUMENT AFTER THE  
ISSUE FEE HAS BEEN PAID (37 CFR 1.55 (a) (2) AND 1.17 (i))**

Sir:

Enclosed is the certified copy of South African Priority Document No. 2002/4112 for the above referenced application. The date of certification is March 11, 2004, and the document is submitted to perfect the applicant's claim for priority made on filing of the application. A check in the amount of \$130.00 is also enclosed in payment of the fee under 37 CFR 1.17(I) for submitting the priority document after the issue fee has been paid.

Respectfully submitted,

  
William J. Sapone  
Registration No. 32,518  
Attorney for Applicant(s)

COLEMAN SUDOL SAPONE, P.C.  
714 Colorado Avenue  
Bridgeport, Connecticut 06605-1601  
Telephone No. (203) 366-3560  
Facsimile No. (203) 335-6779

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*Sertifikaat*

PATENTKANTOOR  
REPUBLIC OF SOUTH AFRICA

DEPARTEMENT VAN  
HANDEL EN NYWERHEID



U.S. 09/927805

*Certificate*

PATENT OFFICE  
REPUBLIEK VAN SUID-AFRIKA

DEPARTMENT OF TRADE  
AND INDUSTRY

Hiermee word gesertifiseer dat  
This is to certify that

the documents attached hereto are true copies of the Forms P2, P6,  
provisional specification and drawing of South African Patent  
Application No. 2002/4112 in the name of Fitter, Johan Christiaan

Filed : 11. August 2000

Entitled : Electrochemical Cells

Geteken te  
Signed at

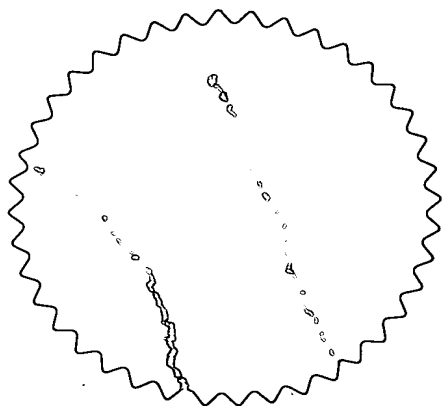
PRETORIA

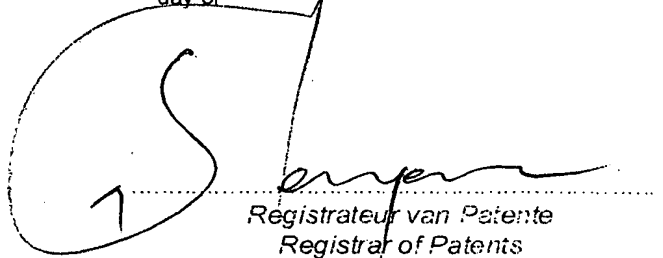
in die Republiek van Suid-Afrika, hierdie  
in the Republic of South Africa, this

11th

dag van  
day of

March 2003



  
Registrateur van Patente  
Registrar of Patents

REPUBLIC OF SOUTH AFRICA		REGISTER OF PATENTS		PATENTS ACT,	
OFFICIAL APPLICATION		LODGING DATE: PROVISIONAL		ACCEPTANCE DATE	
21	01	20004112		22	11-08-00
47					
INTERNATIONAL CLASSIFICATION		LODGING DATE: COMPLETE		GRANTED DATE	
51		23			
FULL NAME(S) OF APPLICANT(S)/PATENTEE(S)					
71	FITTER, Johan Christiaan				
APPLICANTS SUBSTITUTED:				DATE REGISTERED	
71					
ASSIGNEE(S)				DATE REGISTERED	
71					
FULL NAME(S) OF INVENTOR(S)					
72	FITTER, Johan Christiaan				
PRIORITY CLAIMED		COUNTRY		NUMBER	
DATE					
N.B. Use International abbreviation for country (see Schedule 4)		33		31	
				32	
TITLE OF INVENTION					
54	ELECTROCHEMICAL CELLS				
ADDRESS OF APPLICANT(S)/PATENTEE(S)					
51 MOUNT STREET, BRYANSTON, GAUTENG, SOUTH AFRICA					
ADDRESS FOR SERVICE				S AND F REF	
74	SPOOR AND FISHER, SANDTON			JP/F 466/MK/nh	
PATENT OF ADDITION NO.		DATE OF ANY CHANGE			
61					
FRESH APPLICATION BASED ON		DATE OF ANY CHANGE			

REPUBLIC OF SOUTH AFRICA  
PATENTS ACT, 1978  
APPLICATION FOR A PATENT  
AND ACKNOWLEDGEMENT OF RECEIPT  
(Section 30 (1) – Regulation 22)

REPUBLIC OF SOUTH AFRICA FORM P.1  
REVENUE

R 060,00

The granting of a patent is hereby requested by the undermentioned applicant on the basis of the present application filed in duplicate

OFFICIAL APPLICATION NO.

21	01	20004112
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JP/F 466/MK/nh

FULL NAME(S) OF APPLICANT(S)

71	FITTER, Johan Christiaan
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ADDRESS(ES) OF APPLICANT(S)

	51 MOUNT STREET, BRYANSTON, GAUTENG, SOUTH AFRICA
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TITLE OF INVENTION

54	ELECTROCHEMICAL CELLS
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THE APPLICANT CLAIMS PRIORITY AS SET OUT ON THE ACCOMPANYING FORM P.2. THE EARLIEST PRIORITY CLAIM IS:

COUNTRY: NIL	NUMBER: NIL	DATE: NIL
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THIS APPLICATION IS FOR A PATENT OF ADDITION TO PATENT APPLICATION NO.

21	01	
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THIS APPLICATION IS A FRESH APPLICATION IN TERMS OF SECTION 37 AND IS BASED ON APPLICATION NO.

21	01	
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THIS APPLICATION IS ACCOMPANIED BY:

- ☒ 1. A single copy of a provisional specification of 19 pages.  
☒ 2. Drawings of 1 sheet.  
☐ 3. Publication particulars and abstract (Form P.8 in duplicate).  
☐ 4. A copy of Figure of the drawings (if any) for the abstract.  
☐ 5. An assignment of invention.  
☐ 6. Certified priority document(s).  
☐ 7. Translation of the priority document(s).  
☐ 8. An assignment of priority rights.  
☐ 9. A copy of the Form P.2 and the specification of S.A. Patent Application No.  
☐ 10. A declaration and power of attorney on Form P.3.  
☐ 11. Request for ante-dating on Form P.4.  
☐ 12. Request for classification on Form P.9.  
☒ 13. Form P.2 in duplicate.

74	ADDRESS FOR SERVICE: SPOOR AND FISHER, SANDTON
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Dated: 11 August 2000



SPOOR AND FISHER  
PATENT ATTORNEYS FOR THE APPLICANT(S)

REGISTRAR OF PATENTS, DESIGNS, TRADE MARKS AND COPYRIGHT	RECEIVED
2000-08-11	
REGISTRAR OF PATENTS	

REPUBLIC OF SOUTH AFRICA  
PATENTS ACT, 1978

## PROVISIONAL SPECIFICATION

(Section 30(1) -- Regulation 27)

OFFICIAL APPLICATION NO.

LODGING DATE

21	01	20004112
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22	11-08-00
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FULL NAME(S) OF APPLICANT(S)

71	FITTER, Johan Christiaan
----	--------------------------

FULL NAME(S) OF INVENTOR(S)

72	FITTER, Johan Christiaan
----	--------------------------

TITLE OF INVENTION

54	ELECTROCHEMICAL CELLS
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### BACKGROUND OF THE INVENTION

THIS invention relates to electrochemical cells including secondary cells and electroplating cells, and to an additive for use therein.

A disadvantage encountered in an electroplating process concerns the unequal deposition of metal on the surface of the negative electrode. Generally, a higher current density occurs at projections on the surface of the negative electrode leading to a buildup of metal crystals in these areas. This disadvantage is conventionally addressed by the addition of small quantities of smootheners or brighteners to the electrolyte of the electroplating cell. These can include natural gums and resins or synthesized compounds and mixtures which resemble their properties.

Additives have been proposed for zinc-alkaline and lead-acid batteries. For example, a quaternary ammonium polymer was proposed by Rampel, (US Pat No. 3,660,170; May 2, 1972), in respect of a rechargeable battery and an electroplating cell to inhibit the formation of excessive dendritic zinc on the negative plates of these cells.

A variety of quaternary ammonium polymers were proposed by Davis, (US Pat No. 3,877,993; April 15, 1975), for use in dry cells to reduce the stand deterioration of the negative plates of these cells.

A quaternary ammonium monomer was proposed by Lewenstein, (US Pat No. 3,928,066; December 23, 1975), for conventional lead-acid batteries to inhibit the evolution of hydrogen gas from the negative plates and thereby attenuating the loss of water from these batteries.

While significantly modifying the characteristics prevailing at the surfaces of the negative electrodes of these electrochemical cells, none of these prior art measures appear to provide an electrolyte including an additive which is suitable for use interchangeably in different electrochemical cells.

#### SUMMARY OF THE INVENTION

According to the invention there is provided an electrochemical cell comprising opposed positive and negative electrodes, an aqueous electrolyte in ionic contact with the negative electrode, and a deposition inhibiting agent for inhibiting electrodeposition on the negative electrode of metal ions emanating from the positive electrode thereby to reduce or inhibit mass gain of the negative electrode.

The electrochemical cell is preferably a secondary cell, such as a lead acid battery cell, for example, or an electroplating cell, such as a tin electroplating cell, for example. The electrochemical cell may be a cell from a gel electrolyte battery.

The combination of electrolyte and deposition inhibiting agent is preferably interchangeable between the secondary cell and the electroplating cell on an

equivalent volume basis or by concentration or dilution thereof until it reaches the required or optimum specific gravity for use in the secondary cell or electroplating cell, as the case may be.

The deposition inhibiting agent is preferably of a type which is attracted to the surface of the negative electrode, thereby preventing or inhibiting deposition on the negative electrode of metal ions originating from the positive electrode and which are attracted in the normal course to the negative electrode.

The deposition inhibiting agent preferably includes a polar aspect and a non-polar aspect so as to have an affinity for water as well as oils, fats and the like.

In the case of a lead or tin electrode in a sulphuric acid electrolyte, N-alkyl dimethyl benzyl ammonium chloride and sodium dioctyl sulphosuccinate have been shown to be effective to inhibit electrodeposition on these electrodes. Thus, a sulphuric acid electrolyte dosed with either one of these additives can interchangeably be used in a lead acid battery and a tin electro-plating cell.

The invention extends to a method of inhibiting electrodeposition of metal ions on a negative electrode of an electrochemical cell comprising adding to the electrolyte a deposition inhibiting agent which is compatible with the components of the electrochemical cell, non-reactive or slowly reactive with the electrolyte and capable of being attracted to the negative electrode in order to inhibit or prevent deposition thereon of metal ions emanating from the positive electrode which are attracted to the negative electrode.

The invention also extends to an electrochemical cell having a deposition inhibiting agent deposited on a component of the cell operatively in contact with an electrolyte in the cell so that, when an electrolyte is added to the cell, the deposition inhibiting agent associates with the electrolyte so as to be attracted to the negative electrode in order to inhibit or prevent deposition



thereon of metal ions emanating from the positive electrode which are attracted to the negative electrode in use.

#### **BRIEF DESCRIPTION OF THE DRAWINGS**

The invention will now be described in more detail, by way of example only, with reference to the accompanying drawing, which is a schematic representation of an electrochemical cell for evaluating the effect of incorporating an additive of the invention in the electrolyte thereof.

#### **DETAILED DESCRIPTION OF THE INVENTION**

The invention relates to the use of a deposition inhibiting agent or additive with an electrolyte of a type used in an electrochemical cell. The preferred group of chemical compounds suitable as additive is defined as any agent, solid or liquid, which when incorporated into an electrochemical cell electrolyte, promotes a reduction in the rate of deposition of metal on the negative electrodes of the cell, thereby providing an improvement in the quantity and quality of electrodepositioning which is of benefit to the operation of the electrochemical cell.

The type of electrochemical cell suitable for use with an electrolyte and additive of the invention includes battery cells and electroplating cells, as well as other types of cells including electrolytic and electrolysis cells.

In the case of battery cells and some electrolysis cells, the benefit accrues as an increase in the service life of the cells. In the case of electroplating cells and some electrolytic cells, the benefit manifests itself by providing a smoother plated on surface.

Incorporation of the additive into the electrolyte provides the electrolyte with a facility of interchangeability. By way of example, a sulphuric electrolyte including an additive of the invention is suitable for a lead-acid battery cell as well as a tin electroplating cell. The additive of the invention may also be suitable for a gel electrolyte battery cell.

The extent to which interchangeability is effective is such that a quantity of electrolyte incorporating the additive can be drawn from a lead-acid battery cell, through which association it had been providing the benefit accruing to battery cells, for transference to a tin electroplating cell, through which association it will provide the benefit which manifests itself in electroplating cells. In this regard such a receiving electroplating cell is usually not already associated with an additive or an electrolyte at the time of transference.

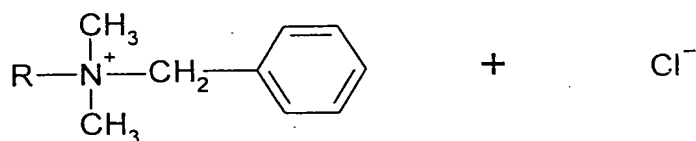
The additive preferably consists of an organic compound that is compatible with the materials of the electrochemical cell and does not react, or reacts only very slowly, with the electrolyte. The organic compound preferably has a polar aspect to its molecule as well as a non-polar aspect, and therefore has an affinity for water as well as oils, fats and the like. The compounds that are successful with a particular metal in combination with a particular electrolyte are not necessarily successful with other metal-electrolyte combinations, and selection is therefore mainly empirical.

The organic compounds that have proved useful with tin and lead electrodes in a sulphuric acid electrolyte include n-alkyl dimethyl benzyl ammonium chloride and sodium dioctyl sulphosuccinate. Significantly the former is cationic and the latter is anionic in solution.

Other compounds that have proved useful to a lesser extent include n-alkyl dimethyl benzyl ammonium sulphate and didecyl dimethyl ammonium chloride. Certain amines, amine salts and amides have also proved useful.

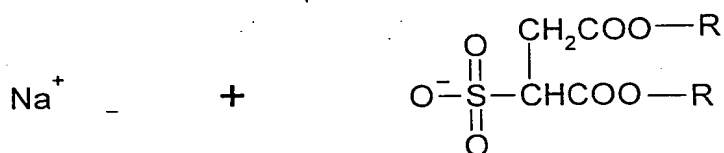
The n-alkyl dimethyl benzyl ammonium chloride is water based while the sodium dioctyl sulphosuccinate is methanol based.

The structure of the n-alkyl dimethyl benzyl ammonium chloride molecule is approximated by the formula:



with R corresponding to the n-alkyl which is a mixture of C<sub>12</sub>, C<sub>14</sub> and C<sub>16</sub>.

The structure of the sodium dioctyl sulphosuccinate is approximated by the formula:

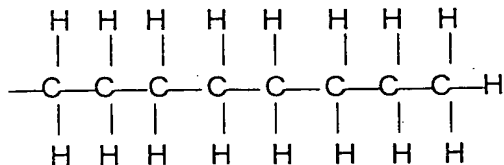


with R corresponding to a C<sub>8</sub> alkyl chain.

Although the particular mechanism of how the agent interacts with the negative electrode of the electrochemical cell is not fully understood, the following is provided as a possible explanation of its effective use.

The charged portion of the molecule as well as the alkyl chains are believed to play important parts in the process which restricts the rate of deposition of metal on the negative electrode of the electrochemical cell.

Alkanes are "straight-chain" hydrocarbons, which consist of an undulating chain of carbon atoms, which in the case of C<sub>8</sub> is represented as follows:



They may be found fully extended, rolled up into a ball or in an intermediate shape. Adjacent molecules typically arrange themselves in a solution in an appropriate shape to maximize the distance between the hydrogen atoms. Although alkyl chains are strongly water repelling, these molecules dissolve in water due to their charged "heads" having an affinity for polar molecules, such as water.

In the case of agents comprising molecules that are ionized, they are expected to be attracted to the surface of the negative electrode by electrostatic attraction in a manner that is similar to the way metal ions are attracted. The degree of attraction is dependent on the electrochemical potential of the substance in question.

It seems reasonable to expect that the charge on the n-alkyl dimethyl benzyl ammonium ion attracts it directly onto the surface of the negative electrode. It is possible that the dioctyl sulphosuccinate ion couples to an intermediary ion present in the electrolyte and the combination is attracted onto the surface of the negative electrode.

Once attached these agents appear to present a web or a tangle of alkyl chains which appear to be comparatively impervious to the metal ions originating from the positive electrode, and which are attracted to the negative electrode.

It further appears from tests carried out that the n-alkyl dimethyl benzyl ammonium ion is attracted to the extremities of metal crystals that are growing

on the surface of the negative electrode and the dioctyl sulphosuccinate ion is attracted closer to the surface of the electrode.

This appears to be borne out by the appearance of the electrode surface after extended operation, in which microscopic examination reveals the crystalline deposition in the case of the n-alkyl dimethyl benzyl ammonium to be significantly blunted in appearance while the dioctyl sulphosuccinate appears to be more stringy.

Despite the difference in appearance, and a risk of the dioctyl sulphosuccinate material becoming dislodged, the deposited material can be weighed, showing that the rate of deposition is, in both cases, very similar.

Referring to Fig 1, an apparatus required to demonstrate the practice of the invention comprises an electrochemical unit 10 which includes a power source 12 and a set of electrochemical cells 14.

The set of electrochemical cells 12 comprises a first cell 16 and a second cell 18. The first cell 16 includes a positive electrode 20, a negative electrode 22 and an electrolyte 24.

The positive electrode 20 and the negative electrode 22 are immersed in, or otherwise conveniently brought into contact with, the electrolyte 24.

The second cell 18 includes a positive electrode 26, a negative electrode 28 and an electrolyte 30. The positive electrode 26 and the negative electrode 28 are immersed in, or otherwise conveniently brought into contact with, the electrolyte 30.

The power source 12 includes a positive output terminal 32 and a negative output terminal 34. The power source 12 provides an electric current of

controlled magnitude to the first cell 16 and the second cell 18, which are connected in series, with the positive terminal 32 connected to the positive electrode 26, the negative electrode 28 connected to the positive electrode 20 and the negative electrode 22 connected to the negative terminal 34. This arrangement ensures that the current flowing through the first cell 16 is matched as closely as is feasible by the current flowing through the second cell 18.

It further ensures that the electrolytic reactions that occur within the first cell 16 and within the second cell 18 may be predicted to be closely matched in accordance with Faraday's laws.

Functionally, an electrochemical cell of the invention, which is not shown, will correspond to the cell 16 and comprises at least one positive electrode corresponding to the electrode 20, at least one negative electrode corresponding to the electrode 22, and an electrolyte which is of the same composition as the electrolyte 24. Typically, the first cell 16 is not provided with an electrolyte at first. Typically, a portion of the electrolyte drawn from the cell of the invention is transferred into the first cell 16, thereby providing the first cell 16 with a requisite quantity of electrolyte for its proper functioning.

Significantly, the positive electrode of the cell of the invention and the corresponding positive electrode 20 need not be constructed of and need not contain the same elements or metals, nor contain any elements or metals they may have in common in an equal proportion.

Similarly, the negative electrode of the cell of the invention and the corresponding negative electrode 22 need not be constructed of and need not contain the same elements or metals, nor contain any elements or metals they may have in common in an equal proportion.

Subsequent to transference of a suitable quantity of the electrolyte drawn from the cell of the apparatus of the invention to the first cell 16, and thereby providing the cell 16 with the requisite electrolyte 24, the second cell 18 is provided with a suitable quantity of an electrolyte 30, which quantity is procured from a source substantially free of any known agent as herein described.

The positive electrodes 20 and 26 typically contain in excess of 90 percent by weight of tin, while the negative electrodes 22 and 28 typically have a 60:40 to a 50:50 tin-lead solder composition.

The positive electrodes 20 and 26 and the negative electrodes 22 and 28 typically consist of wire having a diameter of between 1.6 and 3.2 millimetres, and having a weight of 10 g of which at least 80 percent is submerged.

The electrode wire may be conveniently coiled prior to incorporation into the cells 16 and 18, having a typical coil diameter of 10 millimetres and a spacing between the positive electrode 26 and the negative electrode 28 of at least 30 millimetres. The electrodes 20 and 22 are arranged to match the disposition of the corresponding electrodes 26 and 28.

The electrochemical cells 16 and 18 are each rated to conveniently contain an approximate volume of 100 millilitres of electrolyte. The cells 16 and 18 and their components may be scaled up or down dimensionally, as well as up numerically.

The power source 12 is activated to commence electroplating operation. The current in the circuit is controlled by a regulator represented by a rotatable controller 36. The current in the electrochemical cell circuit is indicated by an ammeter 38 and the voltage across the electrochemical cells 16 and 18 is indicated by a voltmeter 40.

The power source regulates the current in the circuit at 150 milliamperes and the voltage self-adjusts according to cell requirement, but does not exceed 3,5 V/cell. The current is regulated to an accuracy of plus-minus 10 percent.

The use of the above apparatus in determining the efficacy of the invention is set out in the following examples.

#### EXAMPLE 1

A comparison of five samples of electrolyte simulating five samples drawn from various sources was performed. The electrochemical cells 16 and 18 were electrically connected in series and therefore an additional three cells corresponding to the cell 16 and having electrodes corresponding to the electrodes 20 and 22 were connected in series with the cells 16 and 18. While the power source 12 was obliged to supply a voltage totaling  $2\frac{1}{2}$  times the voltage requirement of two cells, the current in the circuit was unchanged.

All five positive electrodes were constructed of 2 millimetre diameter 97:3 tin-silver solder wire and weighed 10.00 g each. All five negative electrodes were constructed of 3.2 millimetre diameter 50:50 tin-lead solder wire and weighed 10.00 g each.

The electrochemical cells were numbered consecutively, with number 1 being allocated to the cell 18, number 2 allocated to the cell 16, and numbers 3, 4 and 5 being allocated to the additional three cells in the circuit.

Measured samples of 100 millilitres of electrolyte were procured from the same container of 1.250 specific-gravity battery-grade sulphuric acid. Each of the five samples received 1.5 millilitres of water containing various additive agents, with exception of electrolyte 1, which received only water.



The 1.5 millilitres of additive raised the volume of electrolyte of all five cells to 101.5 millilitres and reduced the specific gravity marginally but equally.

Subsequent to addition, cell 1 contained plain sulphuric acid in water. Cell 2 contained the same quantity of acid in water plus 300 parts per million, (ppm), n-alkyl dimethyl benzyl ammonium chloride. Cell 3 included 300 ppm n-alkyl dimethyl benzyl ammonium sulphate. Cell 4 included 300 ppm didecyl dimethyl ammonium chloride. Cell 5 included 300 ppm sodium dioctyl sulphosuccinate.

The negative electrodes were weighed after 36 hours of electrochemical activity:

ALL ELECTRODES PRIOR TO TEST	-	10.00 g
NEGATIVE ELECTRODE 1	-	10.99 g
NEGATIVE ELECTRODE 2	-	10.74 g
NEGATIVE ELECTRODE 3	-	10.81 g
NEGATIVE ELECTRODE 4	-	10.95 g
NEGATIVE ELECTRODE 5	-	10.74 g

From the foregoing it can be seen that the plain-electrolyte electrode gained 0.99 g, while the electrodes exposed to additives of the invention gained less weight. The n-alkyl dimethyl benzyl ammonium chloride and the sodium dioctyl sulphosuccinate appear to possess equal efficacy having provided the least weight gain in equal proportion, at roughly 25 percent down.

Since there was some evolution of gas at both the positive and the negative electrodes, it seems reasonable to infer that the differences in the measured weights of the electrodes can be attributed to the chemical equivalent of at least a portion of hydrogen gas emanating from the negative electrodes during the process.

The surface of electrode 1 received a poor quality fluffy covering, electrodes 4 and 5 received a more compact covering and electrodes 2 and 3 received an electroplated finish having potential mirror-quality finish with system adjustment.

## EXAMPLE 2

A comparison of three samples of electrolyte simulating three samples drawn from various sources was conducted. The third cell was accommodated in the test similarly to the way the three extra cells were accommodated for Example 1, with the power source 12 being obliged to supply  $1\frac{1}{2}$  times the voltage requirement of two cells.

All three positive electrodes were constructed out of segments taken out of the positive plates of a motive power lead-acid battery cell. The grid construction contained about 6 percent of antimony in lead. The paste consisted of lead-dioxide. Although it is difficult to match the weights of pasted plate segments upon disassembly of positive battery plates, it was possible to weigh out three plates having a weight of about 15 g each, plus minus 0.5 g.

The rate of electrodeposition of lead on a negative electrode in a sulphuric acid electrolyte is extremely slow. A first test was conducted using negative electrodes identical to those employed in Example 1, but there was no discernible weight gain in any of the negatives after 150 hours.

It is common for lead-acid submarine batteries to be provided with negative electrodes having a grid structure based on copper. This assists in maximizing battery performance during a heavy discharge, as required for attaining maximum speed of the submarine. Copper is suitable for this application since its electrochemical potential, at  $\text{Cu}^{++} + 2\text{e}^- = + 0.34$  volts, which makes it "nobler" than lead at  $\text{Pb}^{++} + 2\text{e}^- = - 0.13$  volts. A plain copper negative can be

introduced into a lead-acid battery cell without discernible deleterious effect provided that a charging current is maintained. A lead film is deposited on the surface of the copper electrode which assists in establishing the potentials pertaining to a conventional lead-acid battery cell.

In the light of the above, negative electrodes were consequently constructed out of a copper foil which possessed a bright reflective surface, and the cells were fully assembled, connected and the power source was energized, before the three samples of electrolyte were poured into all three cells virtually simultaneously.

The electrolyte of cell 6 was provided with ingredients equivalent to the electrolyte of cell 1 in Example 1. The electrolyte of cell 7 was provided with ingredients equivalent to the electrolyte of cell 2 in Example 1. The electrolyte in cell 8 was provided with ingredients equivalent to the electrolyte of cell 5 in Example 1.

The construction of the cells facilitated visual observation of the negative plates undergoing test, and after several hours a visible dulling of the plate surfaces became apparent. This dulling was made easily visible by the reddish colouration and the brightness of the surfaces of the negative electrode plates contrasting sharply against the pronounced greying and dulling produced by the on-plating lead, and antimony, which originated from the positive plates.

After 24 hours the appearance of all three negative plate electrodes had undergone significant change, with electrode 6 appearing quite dark, verging on black with only a slight coppery tint showing; electrode 7 appearing relatively coppery with a slight greyish tinge; and electrode 8 appearing somewhat darker than electrode 7, but very much more coppery than electrode 6.

From these appearances it could be deduced relatively easily that the rate of electrodeposition on electrode 6 had been significantly higher than on electrodes 7 and 8, and that the rate of electrodeposition on electrode 8 had been marginally higher than on electrode 7.

### EXAMPLE 3

Two identical 6 volt type 105 deep cycling lead-acid batteries were subjected to the Battery Council International Deep Cycling Battery Test Procedure (5/93). The tests were carried out at room temperature and spanned a time of approximately seven months. A charging current was applied at 30 amperes, to a voltage limit of 2.55 V/cell, and until 130 percent of the previously discharged capacity was returned. Discharge was applied at 75 amperes, down to a voltage limit of 1.75 V/cell. The batteries were rested for 4 hours before commencing the next cycle.

Battery 1 contained stock standard electrolyte and was replenished every fortnight with plain battery water. Battery 2 contained stock standard electrolyte to which had been added a dosage of n-alkyl dimethyl benzyl ammonium chloride to provide a concentration in the electrolyte of 200 parts per million. Battery 2 was replenished every fortnight with plain battery water to which had been added a dosage of n-alkyl dimethyl benzyl ammonium chloride to provide a concentration in the water of 800 parts per million.

The testing procedure applied to battery 1 and battery 2 was commenced and terminated simultaneously, and the batteries located in close proximity to ensure equalisation of any temperature variations. Battery 1 failed after completing 414 cycles and battery 2 had completed 422 cycles at this point. Both batteries were taken out of the test and were disassembled.

A corresponding cell on each battery was selected, and the elements therein carefully disassembled.

Six negative plates from one cell of battery 1 and six negative plates from the corresponding cell of battery 2 were weighed including their associated separators to ensure that any plated-on material would be included in the measurement:

BATTERY 1 (plain)	BATTERY 2 (additive)
374.4 g	375.0 g
403.8 g	374.5 g
396.8 g	373.4 g
401.2 g	379.6 g
400.0 g	372.4 g
<u>389.4 g</u>	<u>371.8 g</u>
394.3 g (average)	374.5 g (average)

The above batteries were undoubtedly manufactured to exacting standards and their respective plate assemblies can therefore be considered to have almost identical weights at the commencement of the testing procedure. As such the negative plates of the non-treated battery number 1 experienced a plated-on weight gain of about five percent over the negatives of the treated battery number 2.

A third battery of identical type and manufacture, and receiving an equivalent dosing in its replenishment water to that of battery 2 was run concurrently in the test, but was not disassembled. It was functioning correctly at 550 cycles, an increase of approximately 33 percent over the normal expected service life.

#### EXAMPLE 4

A negative electrode identical to the negative electrodes employed in Example 1 was coated with a layer of 50 percent concentration n-alkyl dimethyl benzyl ammonium chloride to an extent approximately equivalent to the portion of the electrode which is submerged in the electrolyte while in use. This equated to about 36 millimetres of length.

The coating procedure included hanging the electrode vertically from a scale and allowing the coating liquid to drip off the electrode subsequent to an excess quantity having been applied. When the electrode weight reached 10.03 g, the electrode was briefly inverted, to allow the coating to establish itself on the surface without further dripping. Thereafter the electrode was baked in an oven at 120°C, (248° F), for two hours.

After this, the electrode was placed in a vessel containing 100 millilitres of water, and the vessel was sealed, and agitated to remove as much as possible of the coating and to dissolve it in the water. Thereafter the concentration of n-alkyl dimethyl benzyl ammonium chloride in the 100 millilitre volume of water was ascertained by spectrophotometer means.

A concentration of 160 parts per million was revealed, which corresponds favourably with a theoretical concentration of a 50 percent solution weighing 0.03 g, at 0.98 SG in 100 millilitres of water, equivalent to 153 parts per million.

There is obviously no weight gain, the difference being due to measurement inaccuracies. However, the procedure does show the feasibility of applying a measured dosage of the additive to any part of an electrochemical cell known to be immersed in the electrolyte in normal use, but to facilitate a suitable delay before application of the electrolyte, when it is more convenient. Thus, an electrochemical cell can be kept in readiness for such application, a simple

addition of electrolyte being the only requisite action to activate the electrolyte-additive combination.

#### EXAMPLE 5

A suitable quantity of electrolyte known to contain a requisite concentration of n-alkyl dimethyl benzyl ammonium chloride, and having a specific gravity of 1.120 was concentrated by simple boiling, until the specific gravity was estimated to have attained a value slightly in excess of 1.250.

Thereafter, upon cooling, water was added until the specific gravity equated to 1.250. A 100 millilitre volume of this adjusted electrolyte was employed in a cell and tested in accordance with the procedure outlined in Example 1. The electrolyte provided substantially identical performance to that of cell number 2.

This simple test demonstrates that an electrolyte can be transferred from, by way of example, a discharged lead-acid battery, to a tin electroplating cell using a simple boiling means to provide concentration to a requisite level. Only water is removed from the electrolyte. The converse is equally valid, and water may be added to adjust the electrolyte as required.

DATED THIS 11<sup>TH</sup> DAY OF AUGUST 2000



SPOOR AND FISHER

APPLICANT'S PATENT ATTORNEYS

